REPORT

TO THE

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

SEMIANNUAL STATUS REPORT #9

for

GRANT NAGW-533

LABORATORY EVALUATION AND APPLICATION OF

MICROWAVE ABSORPTION PROPERTIES UNDER SIMULATED

CONDITIONS FOR PLANETARY ATMOSPHERES

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February 1, 1988 through July 31, 1988

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I. INTRODUCTION AND SUMMARY

Radio absorptivity data for planetary atmospheres obtained from space-craft radio occultation experiments and earth-based radio astronomical obser-vations can be used to infer abundances of microwave absorbing atmospheric constituents in those atmospheres, as long as reliable information regarding the microwave absorbing properties of potential constituents is available. The use of theoretically-derived microwave absorption properties for such atmospheric constituents, or laboratory measurements of such properties under environmental conditions which are significantly different than those of the planetary atmosphere being studied, often leads to significant misinterpreta-tion of available opacity data. Steffes and Eshleman (1981) showed that under environmental conditions corresponding to the middle atmosphere of Venus, the microwave absorption due to atmospheric SO_2 was greater than that calculated from Van Vleck-Weisskopf theory. Similarly, results obtained for the microwave opacity from gaseous H_2SO_4 under simulated Venus conditions, during the first two years of Grant NAGW-533, showed that not only was the opacity from H_2SO_4 much greater than theoretically predicted, but that its frequency (wavelength) dependence was far different than that theoretically predicted (Steffes, 1985 and Steffes, 1986). Measurements made by Steffes and Jenkins (1987), during the third year of Grant NAGW-533, showed that the microwave opacity of gaseous ammonia (NH_3) under simulated Jovian conditions did indeed agree with theoretical predictions to within experimental accuracy at wave-lengths longward of 1.3 cm. Work performed during the fourth year of Grant NAGW-533 (February 1, 1987 through January 31, 1988) and continuing on into the first half of this current grant year (February 1, 1988 through July 31, 1988) has shown that the millimeter-wave opacity of ammonia between 7.5 mm and

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9.3 mm likewise agrees with theoretical predictions. The recognition of the need to make such laboratory measurements of simulated planetary atmospheres over a range of temperatures and pressures which correspond to the altitudes probed by both radio occultation experiments and radio astronomical observations, and over a range of frequencies which correspond to those used in both radio occultation experiments and radio astronomical observations, has led to the development of a facility at Georgia Tech which is capable of making such measurements. It has been the goal of this investigation to conduct such measurements and to apply the results to a wide range of planetary observations, both spacecraft and earth-based, in order to determine the identity and abundance profiles of constituents in those planetary atmospheres.

In some cases, new observations or experiments have been suggested by the results of the laboratory measurements. For example, this facility was initially developed, and then operated, in order to evaluate the microwave absorbing properties of gaseous sulfuric acid (H_2SO_4) under Venus atmospheric conditions. The results, obtained at 13.4 cm and 3.6 cm wavelengths, were applied to measurements from Mariner 5, Mariner 10, and early Pioneer-Venus Radio Occultation experiments, to determine abundances of gaseous sulfuric acid in the Venus atmosphere, with accuracies exceeding those achieved with in-situ instruments (Steffes, 1985). Later efforts concentrated on making laboratory measurements of the microwave absorption from gaseous H_2SO_4 at wavelengths from 1.3 to 22 cm under simulated Venus conditions. We applied these results to radio astronomical observations of Venus which have been made in the same wavelength range, in order to better model the structure of H_2SO_4 and SO_2 abundance in the Venus atmosphere, and to resolve temporal variations of their abundances on a planet-wide basis.

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Our laboratory measurements also suggested that a substantial variation in the Venus microwave emission, related to the abundance of gaseous sulfuric acid, might exist near the 2 cm wavelength. Since no observations of the Venus emission at this wavelength have ever been published, we conducted observations of Venus using the 140-foot NRAO telescope and the 64-meter DSN/Goldstone antenna in April 1987 to not only search for the presence of the predicted feature, but to use such a feature to determine a planet-wide average for sulfuric acid vapor abundance below the main cloud layer. The results of this observation are substantial in that they not only place limits on the abundance and spatial distribution of gaseous H_2SO_4 and SO_2, but they also suggest some limits to long term temporal variations for the abundance of these two gases. During the first half of this current grant year, we have completed calibration and interpretive studies on the data from these observations and are submitting a paper entitled, "Observations of the Microwave Emission of Venus from 1.3 to 3.6 cm," by P. G. Steffes, M. J. Klein, and J. M. Jenkins, to the journal <u>Icarus</u>. (Preprints of this paper will accompany this report under separate cover.)

Another important tool for evaluating potential spatial and temporal variations in abundance and distribution of gaseous H_2SO_4 is the reduction and analysis of recently obtained Pioneer-Venus radio occultation measurements. The 13 cm microwave absorptivity profiles, which can be obtained from the radio occultation data, are closely related to the abundance profiles for gaseous H_2SO_4. Within the last month, we have begun the reduction of the 1986-87 Pioneer-Venus radio occultation measurements (working at JPL with support from the Pioneer-Venus Guest Investigator Program) in order to obtain the needed 13 cm microwave absorptivity profiles. This reduction effort, and its potential results, are discussed in Section V of this report.

An equally important activity for the first half of this grant year has continued to be laboratory measurements of the microwave and millimeter-wave properties of the simulated atmospheres of the outer planets and their satellites. As described in the previous Annual Status Report for Grant NAGW-533 (February 1, 1987 through January 31, 1988), initial laboratory measurements of the millimeter-wave opacity of gaseous ammonia (NH_3) in a hydrogen/helium (H_2/He) atmosphere, under simulated conditions for the outer planets were begun in 1987. These measurements were conducted at frequencies from 32 to 40 GHz (wavelengths from 7.5 to 9.3 mm). It has been found by some (e.g., de Pater and Massie, 1985) that the observed millimeter-wave emission from Jupiter is inconsistent with the millimeter-wave absorption spectrum predicted using the modified Ben-Reuven line shape for ammonia. In order to investigate this, we developed a Fabry-Perot spectrometer system capable of operation from 30 to 41 GHz (wavelengths from 7.3 to 10 mm). This system has been used at pressures up to 2 Bars and temperatures as low as 150 K, which corresponds closely to the conditions at altitudes in the Jovian atmosphere most responsible for the observed millimeter-wave absorption. A complete description of the millimeter-wave spectrometer is given in Section II.

Initially, we used this spectrometer to complete laboratory measurements of the 7.5 to 9.3 mm absorption spectrum of ammonia. The results of these measurements were substantive in that they suggested the possibility that neither the modified Ben-Reuven lineshape nor the Van Vleck-Weisskopf lineshape best describe the 7.5 to 9.3 mm (32 to 40 GHz) absorption from gaseous NH_3 under simulated Jovian conditions. However, because of the large error bars for these initial measurements, it was not possible to determine the specific absorption spectrum. In order to resolve this uncertainty, we

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have found that it is desirable to characterize the opacity of ammonia to an accuracy of ±20%. This would also help to determine upper level ammonia abundance and distribution in the atmospheres of the outer planets (from radio emission measurements), and to properly account for contributions to opacity from other absorbing constituents. For our initial measurements, accuracies of no better than ±60% were achieved (see Joiner et al., 1987). Thus, we have devoted a great deal of effort during the first half of this current grant year to improve the sensitivity of our 7.5 to 10 mm spectrometer system, as described in Section II. The effect of this improvement can be seen in the laboratory results described in Section IV.

Since larger variations from theoretically-derived opacity values are expected at shorter millimeter-wavelengths (see de Pater and Massie, 1985), we hope to pursue (in the second half of this grant year) further laboratory measurements at shorter millimeter-wavelengths, especially near 3.2 mm (94 GHz), where a large number of observations of the emission from the outer planets have been made. A better knowledge of the millimeter-wave absorption properties of NH_3 is essential, not only to help better characterize the distribution and abundance of ammonia at high levels in Jovian atmospheres, but to make it possible to resolve the contributions from other absorbing constituents such as H_2S (see Bezard et al., 1983).

Of equal importance, however, will be the further application of our laboratory results for the microwave and millimeter-wave absorption from various gases under simulated planetary conditions to available absorptivity data. These data sets include microwave and millimeter-wave emission measurements from Venus and the outer planets, from which opacity can be inferred. Other sources of absorptivity data include the Pioneer-Venus radio

occultation experiments, discussed in Section V, and Voyager radio occultation experiments at the outer planets. Results of our laboratory measurements have also been used in the study and planning of future missions (see Section VI).

II. THE GEORGIA TECH RADIO ASTRONOMY AND PROPAGATION (R.A.P.) FACILITY

The basic configuration of the planetary atmospheres simulator developed at Georgia Tech for use in measurement of the microwave absorptivity of gases under simulated conditions for planetary atmospheres is described at length in the previous Annual Status Report(s) for Grant NAGW-533. It is also discussed at length in Steffes (1986) and Steffes and Jenkins (1987). The most recent addition to the Georgia Tech Radio Astronomy and Propagation Facility has been a Fabry-Perot type resonator capable of operation between 30 and 41 GHz. As shown in Figure 1, the resonator consists of two gold plated mirrors (one with a flat surface, and one with a parabolic surface) separated by a distance of about 20 cm. The mirrors are contained in a T-shaped glass pipe which serves as a pressure vessel capable of withstanding over 2 atm of pressure. Each of the three open ends of the pipe is sealed with an O-ring sandwiched between the lip of the glass and a flat brass plate which is bolted to an inner flange. Electromagnetic energy is coupled both to and from the resonator by twin irises located on the surface of the flat mirror. Two sections of WR-28 waveguide which are attached to the irises pass through the brass plate to the exterior of the pressure vessel. The end of each waveguide section is pressure-sealed by a rectangular piece of mica which is held in place by a mixture of rosin and beeswax. As shown in Figure 2, one of these ends is connected to the sweep oscillator through a waveguide section. A Ka-band (26-40 GHz) mixer is attached to the other section of waveguide and is coupled to

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the high resolution spectrum analyzer through a calibrated section of coaxial cable. The entire resonator, including its glass pressure envelope, is placed in the temperature chamber, which is a low-temperature freezer capable of operation down to 150 K. A network of stainless steel tubing and valves connects other components such as gas storage tanks, vacuum gauges, the pressure gauge, and the vacuum pump to the resonator assembly, so that each component may be isolated from the system as necessary. When properly secured, the system is capable of containing up to two atmospheres of pressure without detectable leakage. The sensitivities (minimum detectable opacities) previously achievable with this system are shown in Figure 3 (solid line). However, as previously discussed, these minimum sensitivities were too large to allow an unambiguous determination of the absorption spectrum of ammonia under simulated Jovian conditions.

In order to achieve a better system sensitivity, which corresponds to a higher "Q" or quality factor for the Fabry-Perot resonator (see Section III), all losses in the resonator must be minimized, since the quality factor is defined as 2π times the ratio of the average energy stored in the resonator to the energy lost (per cycle) in the resonator. There are three sources of loss which typically affect a Fabry-Perot resonator (Collin, 1966):

(1) Resistive losses on the surfaces of the mirrors.

(2) Coupling losses due to the energy coupling out of the resonator through the irises on the flat-surfaced mirror.

(3) Diffraction losses around the sides of the mirrors.

For previous measurements made at frequencies below 22 GHz (wavelengths longer than 1.35 cm), it was found that the resistive losses were predominant. This was because the measurements were conducted using cylindrical resonators,

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for which no diffraction losses existed. (See Steffes and Jenkins, 1987.) Also, coupling losses were held to a minimum by using very small coupling loops and irises. The predominance of the resistive losses in the cylindrical resonators was demonstrated when such resonators were cooled to 193 K in the atmospheric simulator. Significant improvement in the quality factor of the resonators were observed when compared with their room temperature values. This was consistent with the expected reduction in the resistive losses at lower temperatures.

When the newer, higher frequency (30 to 41 GHz) Fabry-Perot resonator (Figure 1) was first cooled from room temperature down to 203 K for tests under simulated Jovian conditions, its quality factor appeared to worsen rather than improve. Initially, it was thought that this might have been caused by separation of the gold plating on the mirror surfaces from the backstructure (which had been machined from aluminum) due to differential thermal contraction. As a result, new mirrors were machined (to high tolerance) from brass, and then were plated with titanium and then gold, to assure no separation would occur. The performance of the new mirrors was only marginally better when installed in the resonator. Computation of the resistive losses from the mirrors showed that, in the absence of all other losses, the Q of our Fabry-Perot resonator should be on the order of 250,000; whereas its actual Q was on the order of 10,000. Therefore, it became clear that either coupling losses or diffraction losses were the limiting factor in its performance, and that even the introduction of high-temperature superconducting material would not significantly improve the sensitivity of the system. (Note, however, that we are still studying the possibility of using high temperature superconductors in our lower frequency, cylindrical resonators in order to obtain increased sensitivity at frequencies below 22 GHz.)

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In order to further improve the quality factor of the 30 to 41 GHz system, some additional improvements were made. First, adjustable irises were developed so that the smallest possible coupling losses would occur, while still allowing sufficient signal coupling in and out of the resonator so as to make accurate absorptivity measurements. Since the irises are actually circular holes which are placed near the center of the flat-surfaced mirror, adjustment of their sizes is difficult. However, two small metal sheets with V-shaped cuts were placed immediately behind each iris in an area where the mirror surface thickness is very small. The two sheets could be moved together or apart so as to adjust the effective size, and therefore the coupling, of each iris. Even when adjusted for minimal coupling, however, the resonator Q was only slightly improved, suggesting that diffraction losses were the major limiting factor to system sensitivity.

Diffraction losses occur due to energy being lost around the edges of the mirrors. These losses can be minimized by assuring that both mirrors are oriented directly toward each other (i.e., the centerlines for each mirror, which are orthogonal to the planes of each mirror at their centerpoints, must be colinear). Since the positioning of the mirrors can vary with temperature, due to thermal contraction or expansion of metallic mounting structures, the temperature dependence of the quality factor which has been observed is consistent with a system which is limited by diffraction losses.

Two approaches can be used to minimize diffraction losses. The first involves precise pointing of the mirrors. This was accomplished by directing the beam of a helium-neon laser through the input waveguide and iris and into the resonator. Mirror positioner screws could then be adjusted so that the reflected beam focused precisely on the output iris. Since the parabolic

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mirror has a precisely defined focus, adjustment of its exact position is far more critical than that of the flat mirror. The second technique for reducing diffraction loss involves the use of larger mirrors in the resonator. However, because of size limitations set by our pressure vessel, we are unable to significantly increase the size of the mirrors in our system.

Overall, our efforts during the first half of this grant year at improving the quality factor of our 30 to 41 GHz Fabry-Perot resonator have been successful, but in themselves have not been enough to provide the needed increase in system sensitivity. However, since absorptivity is measured by monitoring the change in the quality factor of the resonator which is caused by the absorbing gas mixture, improvements in our measurement technique, described in Section III, have allowed us to achieve the required system sensitivity.

III. EXPERIMENTAL APPROACH

The approach used to measure the microwave absorptivity of test gases in an H_2/He atmosphere is similar to that used previously by Steffes and Jenkins (1987) for simulated Jovian atmospheres. At frequencies between 30 and 41 GHz, the changes in the Q of the numerous resonances of the Fabry-Perot resonator (see Figure 2) are related to the absorptivity of the test gas mixture at these frequencies. The changes in the Q of the resonances which are induced by the introduction of an absorbing gas mixture can be monitored by the high resolution microwave spectrum analyzer, since Q is simply the ratio of the cavity resonant frequency to its half-power bandwidth. For relatively low-loss gas mixtures, the relation between the absorptivity of the gas mixture and its effect on the Q of a resonance is straightforward:

$$\alpha \simeq \left(Q_L^{-1} - Q_C^{-1}\right)\pi/\lambda \qquad (1)$$

where α is absorptivity of the gas mixture in Nepers km^{-1}. (Note, for example, that an attenuation constant or absorption coefficient or absorptivity of 1 Neper km^{-1} = 2 optical depths per km (or km^{-1}) = 8.686 dB km^{-1}, where the first notation is the natural form used in electrical engineering, the second is the usual form in physics and astronomy, and the third is the common (logarithmic) form. The third form is often used in order to avoid a possible factor-of-two ambiguity in meaning.) Q_L is the quality factor of the cavity resonator when the gas mixture is present, Q_C is the quality factor of the resonance in a vacuum, and λ is the wavelength (in km) of the test signal in the gas mixture.

In the first half of this grant year, we have attempted to make high accuracy measurements of the millimeter-wave absorption from gaseous ammonia (NH_3) in a 90% H_2/10% He atmosphere at a temperature of 203 K. While even lower temperatures could be achieved, the need to avoid the risk of ammonia condensation kept our operating temperatures relatively high. As in the previous experiments, the bandwidth and center frequencies of each of several resonances between 32 and 41 GHz were measured in a vacuum. Next, 28 torr of gaseous ammonia is added to the system. The pressure of the ammonia gas is measured with the high-accuracy thermocouple vacuum gauge, as shown in Figure 2.

In addition, the ammonia abundance can be monitored by measuring refractivity of the introduced gas. Since the index of refraction (relative to unity) is proportional to the ammonia gas abundance, the ability of the system

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to accurately measure refractivity (through measurement of the frequency shift of resonances) can be used to infer the relative vapor abundance or pressure. Note that it is not yet possible to use this approach for the accurate determination of absolute NH_3 pressure since accurate refractivity data for the 7.3 to 10 mm wavelength range is not available. (In fact, by using our thermocouple vacuum gauge, we have made measurements of the density-normalized refractivity of gaseous ammonia at 39 GHz, and found it to be 8.8 x 10^{-17} N-units/molecule/cm, which is nearly 8 times the value at optical wavelengths.)

Next, 1.8 atm of hydrogen (H_2) and 0.2 atm of helium (He) are added to the chamber, bringing the total pressure to 2 atm. The bandwidth of each resonance is then measured and compared with its value when the chamber was evacuated in order to determine the absorptivity of the gas mixture at 2 atm total pressure. The total pressure is then reduced, by venting, to 1 atm, and the bandwidths are again measured. Finally, the pressure vessel is again evacuated and the bandwidths again measured so as to assure no variation of the Q's of the evacuated resonator has occurred. As with the previous measurements, the measured changes of bandwidths (Q's) can then be used to compute the absorptivity of the gas mixtures at each of the resonant frequencies.

This approach has the advantage that the same gas mixture is used for the absorptivity measurements at the various pressures. Thus, even though some small uncertainty may exist as to the mixing ratio of the initial mixture, the mixing ratios at all pressures are the same, and thus the uncertainty for any derived pressure dependence is due only to the accuracy limits of the absorptivity measurements, and not to uncertainty in the mixing ratio. (This

assumes that the mixing ratio is small, so that foreign-gas broadening predominates, as is the case for our measurements.) Similarly, measurements of the frequency dependence of the absorptivity from the mixture would likewise be immune to any mixing ratio uncertainty, since foreign-gas broadening predominates.

For the measurements described, the amount of absorption being measured is extremely small. Thus, any errors in measurements of (or other changes in) the apparent bandwidth of the resonances, not caused by the absorbing gases, could lead to significant errors in the absorption measurement. The contribution of instrumental errors and noise-induced errors on such absorptivity measurements have been discussed at length in Steffes and Jenkins (1987). However, because our latest measurements represent such small percentage changes in bandwidth, another instrumental source of error which we refer to as dielectric loading becomes a concern.

As can be seen in Figure 2, the resonator, which operates as a bandpass filter, is connected to a signal source (the millimeter-wave sweep oscillator) and to a signal receiver (the high-resolution spectrum analyzer). The "Q" of the resonator, which is defined as the ratio of energy stored in the resonator to the energy lost per cycle, equals the ratio of resonant center frequency to resonance half-power bandwidth. It is not surprising, therefore, that the stronger the coupling between the resonator and the spectrum analyzer or sweep oscillator, the lower will be the Q of the resonance, since more energy will be lost per cycle through the waveguides connecting the resonator to the spectrum analyzer and sweep oscillator. For this reason, we have always designed our resonators (both the coaxially-coupled cylindrical cavity resonators used below 25 GHz and the waveguide-coupled Fabry-Perot resonator

used above 30 GHz) with minimal coupling, so as to maximize Q and to minimize the changes in Q that might result from changes in coupling that occur when gases are introduced into the resonators. It should be noted that these changes in coupling, which are due to the presence of the test gas mixtures, are not related to the absorptivity of the gases, but rather to the dielectric constant or permittivity of the test gas mixtures. (Hence, the term "dielectric loading.")

We have always strived to design the coupling elements of the resonators so that the changes in lossless test gas abundances (and resulting changes in dielectric constant) had little or no effect on the Q of the resonator as measured in the system. This has been no small feat in that slight imperfections in resonators, cables, coupling loops, or waveguides can make the apparent Q of the resonator appear to vary with the abundance of such lossless gases. It has now become a standard part of our experimental procedure to repeat absorption measurements for gas mixtures in which the absorbing gas is a minor constituent, without the absorbing gas present. For example, after measurements were made of the microwave and millimeter-wave absorption from ammonia as a minor constituent in an H_2/He atmosphere, measurements of the apparent absorption of the H_2/He atmosphere without the ammonia gas were made. Since, for the pressures and wavelengths involved, the H_2/He atmosphere is essentially transparent, no absorption was expected. If any apparent absorption was detected, "dielectric loading," or a change in coupling due to the dielectric properties of the gases, was indicated.

Initially, if any evidence of dielectric loading existed, the experiments were terminated and the apparatus disassembled, including pressure seals. The cables and coupling loops were then readjusted, and the system reassembled and

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tested again. The entire procedure was repeated until the dielectric loading effect was eliminated or minimized. If some small variation in the resonant Q or bandwidth due to the presence of the non-absorbing gases still remained, it was added to the uncertainty or error bars for each experiment. More recently however, we have found that the effects of dielectric loading are additive, in that they add to the apparent changes of resonator bandwidth caused by the absorbing gases. Thus, as long as the effects of dielectric loading are not time variable, they can be removed by using the measured value of the Q of a resonance with the non-absorbing gases present (instead of the Q of the resonance in a vacuum) for the quantity Q_c in equation (1).

Another potential source of instrumental error which we have recently detected has to do with nonlinearities in the spectrum analyzer display. We have found that depending on the vertical position of the bandpass spectrum on the spectrum analyzer CRT display, the peak signal level (and, therefore, the apparent half-power bandwidth and resulting quality factor) of the resonator seems to vary slightly. This is due to nonlinearities in the CRT vertical deflection amplifier. We have minimized this potential error by always resetting the vertical position of the displayed spectrum to the same portion of the CRT screen.

As previously discussed, once a test gas mixture is formed in the pressure vessel, the same mixture is used for measurement of absorption at several frequencies and pressures. Thus, even though some uncertainty in absolute mixing ratio exists, the pressure and frequency dependences of the millimeter-wave absorption can be measured to high accuracy. However, in order to properly characterize the magnitude of the absorption, the ammonia mixing ratio must be known precisely. Using the high accuracy thermocouple

vacuum gauge shown in Figure 2, the actual NH_3 mixing ratio can only be determined to an accuracy of ±20% of its value. (Note: This corresponds to (1.85 ± 0.37)% NH_3 volume mixing ratio.) However, since our required overall accuracy for the NH_3 absorptivity measurement is ±20%, this mixing ratio uncertainty is excessive. In order to reduce this uncertainty, we arranged for a local gas products supplier (Matheson Gas Products) to provide us with a pre-mixed hydrogen/helium/ammonia atmosphere which was analyzed with a mass spectrometer so that mixing ratio accuracies of better than 2% (i.e., (1.85 ±.04)%) were obtained. We have used this mixture (1.85% NH_3, 9.81% He, and 88.34% H_2) for the high accuracy absorptivity measurements which are required to accurately infer ammonia abundance from millimeter-wave opacity data for the Jovian planets.

IV. RESULTS OF LABORATORY MEASUREMENTS AND THEIR APPLICATION

Initial measurements of the 7.5 to 9.2 mm absorptivity from NH_3 in a hydrogen/helium atmosphere were conducted at 203 K as described in Section III, with an ammonia mixing ratio of 0.0186, at pressures of 1 and 2 Bars. An examination of these early experimental results revealed that because of large error bars, we could not determine whether the modified Ben-Reuven lineshape best described the absorption profile of gaseous ammonia shortward of 1 cm, as discussed in Sections I and II. As a result, we have undertaken higher accuracy measurements in the first half of this current grant year.

Results of measurements of the 32 to 40 GHz (7.5 to 9.2 mm) absorptivity of gaseous ammonia under simulated Jovian conditions (203 K) are shown in Figure 4. These measurements were made using a 88.34% hydrogen/9.81% helium

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atmosphere with a total pressure of 2 Bars. The ammonia mixing ratio was 0.0185. Triangular points represent measurements of gas mixtures formed using the thermocouple vacuum gauge (NH_3 mixing ratio accuracy = ±20% of its value) and the circular points represent measurements of the pre-mixed, analyzed gas mixture described in Section III. With this mixing ratio, temperatures as low as 190 K could have been used before saturation would have become a problem, but 203 K was used so as to be consistent with earlier measurements.

Also, shown in Figure 4 are solid lines which represent the theoretically-computed opacity using the Van Vleck-Weisskopf lineshape (upper line), the modified Ben-Reuven lineshape (middle line), and the Zhevakin-Naumov lineshape (lower line). The Van Vleck-Weisskopf calculation was performed using linewidths and line intensities as per Wrixon et al. (1971). The Ben-Reuven calculation was made as per Berge and Gulkis (1976), by employing a Ben-Reuven lineshape which has been modified so as to be consistent with the laboratory results of Morris and Parsons (1970), in which the 9.58 GHz absorption from NH_3 (in a high pressure H_2/He atmosphere) at room temperature was measured. The Zhevakin-Naumov calculation used the lineshape of Zhevakin and Naumov (1967) and linewidths and line intensities from Wrixon et al. (1971). These theoretical spectra were computed using generalized computer programs for which the partial pressures from H_2, He, and NH_3, as well as frequency and temperature, were adjustable variables. The values picked for these variables matched our experimental conditions.

Inspection of the results in Figure 4 shows that most of the measured data points lie nearest to the theoretically-derived absorptivity expression based on the Zhevakin-Naumov lineshape. This differs from our preliminary measurements which suggested that the ammonia opacity might actually exceed

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that indicated by the modified Ben-Reuven lineshape. Such results are not surprising, however, in that the accuracy of the new measurements is far greater than that from previous measurements. It is also noteworthy that other researchers, such as de Pater and Massie (1985), have found that in order to best explain the 1-10 mm Jupiter emission spectrum, a different sort of lineshape was needed to characterize the ammonia opacity. Likewise, previous laboratory measurements of NH_3 opacity at frequencies below 22 GHz showed opacities less than those indicated by the modified Ben-Reuven lineshape under the same conditions of temperature and pressure (Steffes and Jenkins, 1987). Since even larger variations from either the modified Ben Reuven formulation or the Van Vleck-Weisskopf formulation for ammonia opacity are expected at shorter millimeter-wavelengths, we hope to pursue further laboratory measurements, especially near 3.2 mm (94 GHz), where a larger number of observations of the emission from the outer planets have been made.

A better knowledge of the millimeter-wave absorption properties of NH_3 is essential, not only to help better characterize the distribution and abundance of ammonia at high levels in Jovian atmospheres, but to make it possible to resolve the contributions from other absorbing constituents such as H_2S (see Bezard et al., 1983). Our goal to better characterize the millimeter-wave absorption spectrum of ammonia will not only involve increasing the range of frequencies over which measurements are made, but to increase the sensitivity of the measuring systems.

V. OBSERVATIONAL AND INTERPRETIVE STUDIES

As described in the previous Annual Status Report for Grant NAGW-533 (February 1, 1987 through January 31, 1988), studies of our recent measure-

ments of the 1.35 to 3.6 cm emission from Venus have suggested that long term temporal and/or significant spatial variations in the abundances of SO_2 and gaseous H_2SO_4 may occur immediately below the main cloud layer (48 km and below). Our observation, which was predominantly of equatorial and mid-latitude regions of Venus, indicated a significantly lower SO_2 abundance than was measured in 1978 by the Pioneer-Venus Sounder Probe, and a lower average abundance of gaseous H_2SO_4 than would have been inferred from earlier Pioneer-Venus radio occultation studies of subcloud opacity at 13 cm. Some or all of this difference may be due to spatial variations in the subcloud H_2SO_4 abundance since most of the early Pioneer-Venus results were for polar latitudes (Cimino, 1982). Similarly, our results may be consistent with the earlier equatorial 13 cm radio occultation opacity measurements made with the Mariner 10 spacecraft (Lipa and Tyler, 1979), where the peak opacity would correspond to a very large abundance of gaseous H_2SO_4, but the average subcloud opacity was significantly lower.

One important tool for evaluating these effects is the reduction of the microwave data from the 1986-87 Pioneer-Venus radio occultation measurements. This data was taken over a wide range of latitudes and could be critical for determining whether temporal variations or spatial variations in gaseous H_2SO_4 abundance could be occurring. Working with Dr. Arvydas J. Kliore (P-V Radioscience Leader), our group has obtained the currently unreduced data and (working at JPL) has begun reducing the data to obtain absorptivity profiles for the 1986-87 epoch.

Over the next year, we will reduce data obtained from the Fall 1986/ Winter 1987 Pioneer-Venus Radio Occultation Observations. Since the initial conversion from amplitude and doppler (frequency) data to refractivity and

absorptivity profiles can be most efficiently completed at JPL, we have made arrangements to send graduate students to JPL for this activity. Support for travel and student salaries for the reduction effort at JPL has been obtained from the Pioneer-Venus Guest Investigator Program.

Since the overall effort is of limited scope, we have selected a small number of radio occultations which are spread over a range of latitudes from polar to equatorial. We will correct the "raw" radio occultation data (amplitude and doppler) for spacecraft motions, frequency drift, and for antenna pointing inaccuracies, in order to then invert the data to obtain reliable refractivity and absorptivity profiles. Since the Pioneer-Venus Orbiter can no longer be positioned to track the planetary limb during occultations and because of spacecraft "wobble," use of the 3.6 cm wavelength has been extremely limited. This is not a severe constraint, however, since under optimal conditions the 3.6 cm wavelength can probe no deeper than 50 km altitude (Cimino, 1982), and we are most interested in altitudes below 48 km.

After the initial reduction, we hope to have dependable 13 cm wavelength refraction and absorption profiles for a range of altitudes in the Venus atmosphere reaching down to 38 km and for latitudes ranging from equatorial to polar. Figure 5 shows a preliminary 13 cm absorptivity profile derived from radio data obtained during the entry occultation of Orbit 2801 on August 6, 1986. (Note: This is the first such absorptivity profile which has been derived since Orbit 358 - November 28, 1979.) This occultation probed the Venus atmosphere at 52°N latitude and the ray path traversed mainly the night side of the planet. For this orbit, the spacecraft signal was only receivable at the Goldstone DSS-14 receiver down to a periapsis altitude of 43.7 km before receiver lock was lost. (This assumes a planetary radius of 6052 km.)

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It is hoped that other orbits may be found which probe deeper into the atmosphere before loss of signal, but because the Pioneer-Venus steerable antenna no longer tracks the limb of the planet (the direction the radio ray travels back to earth) during the occultation, the resulting lower signal level may prevent probing deeper than the 40 km altitude.

To show the usefulness of the 13 cm opacity data for inferring the nature of the gaseous H_2SO_4 abundance, we compare in Figure 6 the measured absorptivity profile from orbit 2801N with that absorptivity which would result from a saturation abundance of gaseous H_2SO_4 (from Steffes, 1985) in the 43 to 55 km altitude range. It can be seen that for altitudes in the 49 to 51 km altitude range (the nominal altitude range of the Venus lower cloud – see Ragent and Blamont, 1980), absorptivity values close to those caused by a saturation abundance of gaseous H_2SO_4 are seen. At lower altitudes, most values for absorptivity are below those caused by a saturation abundance. It should be noted that error bars for this preliminary absorptivity data have not yet been computed, but are expected to be on the order of ± 0.001 dB/km (± 0.00023 km^{-1}).

Figure 7 shows the full extent to which application of our laboratory results can be carried. The abundance of gaseous H_2SO_4 (derived from the absorptivity profile in Figure 5 by using laboratory results from Steffes, 1985) is plotted as a function of altitude, along with a plot of the saturation abundance of gaseous H_2SO_4, for comparison. As our work in the Pioneer-Venus Guest Investigator Program yields more absorptivity profiles for a wide range of locations in the Venus atmosphere, we hope to be able to well characterize the abundance, structure, and spatial variations of gaseous H_2SO_4 in the Venus atmosphere. We also hope to make comparative studies with earlier

radio occultation measurements in order to detect possible temporal variations in H_2SO_4 abundance and structure.

VI. PUBLICATIONS AND INTERACTION WITH OTHER INVESTIGATORS

In the first half of the current grant year, a paper was completed and accepted for publication in Icarus, describing results and applications of some experiments performed during the previous year of Grant NAGW-533 (Jenkins and Steffes, 1988). This paper describes laboratory measurements of the microwave absorption of methane (CH_4) and water vapor (H_2O) under simulated Jovian conditions. The paper also concludes, that based on these laboratory results, neither methane nor water vapor can be responsible for the excess microwave opacity detected at wavelengths between 10 and 20 cm in the atmosphere of Jupiter. This supports the presence of an ammonia abundance which exceeds solar abundance by a factor of 1.5 in the 2 to 6 Bar levels in Jupiter's atmosphere.

In addition, as discussed in Section I, we have just completed a paper describing observations and interpretive studies of the 1.3 to 3.6 cm Venus emission spectrum (Steffes et al., 1988). Likewise, a paper describing the results and applications of the laboratory measurements of the millimeter-wave opacity of ammonia described in Section IV is currently in preparation. We also submitted updated summaries of our most recent laboratory measurements for inclusion in the twenty-first issue of the Newsletter of Laboratory Spectroscopy for Planetary Science.

In addition to the observations of Venus and analysis work conducted jointly with Dr. Michael J. Klein of JPL, we have also worked with Dr. Michael

A. Janssen of JPL regarding models for the Venus atmosphere, interpretation of microwave emission measurements, and theoretical models for the absorption spectrum of H_2SO_4. We have also worked with Dr. Arvydas J. Kliore of JPL on the reduction and interpretation of data from recent Pioneer-Venus Radio Occultation Studies as part of our involvement in the Pioneer-Venus Guest Investigator Program. More informal contacts have been maintained with groups at the California Institute of Technology (Dr. Duane O. Muhleman and his students, regarding interpretation of radio astronomical measurements of Venus opacity), at the Stanford Center for Radar Astronomy (V. R. Eshleman, G. L. Tyler, and T. Spilker, regarding Voyager results for the outer planets, and laboratory measurements), and at JPL (Drs. Robert Poynter and Samuel Gulkis, regarding radio astronomical observations of the outer planets and Venus). We have also worked with Dr. Imke de Pater (University of California-Berkeley) by using our laboratory measurements of atmospheric gases in the interpretation of radio astronomical observations of Venus and the outer planets. We have also studied possible effects of the microwave opacity of cloud layers in the outer planets' atmospheres. In this area, we have worked both with Dr. de Pater and with Dr. Paul Romani (NRC Associate, Goddard SFC). Dr. Steffes has also been active in the review of proposals submitted to the Planetary Atmospheres Program at NASA (both as a "by-mail" reviewer and as a panel member) and as a reviewer of manuscripts submitted to Icarus and the Journal of Geophysical Research, for which Dr. Steffes is an Associate Editor. We have also continued to serve the planetary community through the distribution of reprints of our articles describing our laboratory measurements and their application to microwave and millimeter-wave data from planetary atmospheres. The results of these measurements have been used in the

mission planning for radio and radar systems aboard the Galileo and Magellan missions, and more recently, for proposed experiments for the Cassini mission. Dr. Steffes also participated as a member of the International Jupiter Watch (IJW) Laboratory/ Theory Discipline Team. Another source of close interaction with other planetary atmospheres principal investigators has been Dr. Steffes' membership in the Planetary Atmospheres Management and Operations Working Group (PAMOWG). Travel support for attendance at PAMOWG meetings has been provided by Georgia Tech in support of Planetary Atmospheres Research. Also in support of Planetary Atmospheres Research, Georgia Tech provided $2,000 for required repairs and maintenance to the ultra-cold freezer system used in the outer planets atmospheric simulator.

VII. CONCLUSION

In the first half of this grant year, we have continued to conduct laboratory measurements of the millimeter-wave properties of atmospheric gases under simulated conditions for the outer planets. Significant improvements in our current system have made it possible to accurately characterize the opacity from gaseous NH_3 at longer millimeter-wavelengths (7 to 10 mm) under simulated Jovian conditions. In the second half of this grant year, we hope to extend such measurements to even shorter millimeter-wavelengths. We will likewise continue to pursue further analysis and application of our laboratory results to microwave and millimeter-wave absorption data for the outer planets, such as results from Voyager Radio Occultation experiments and earth-based radio astronomical observations. We also intend to pursue the analysis of available multi-spectral microwave opacity data from Venus, including data from our most recent radio astronomical observations in the 1.3 to 3.6 cm

wavelength range and newly obtained Pioneer-Venus Radio Occultation measure-ments at 13 cm, using our laboratory measurements as an interpretive tool.

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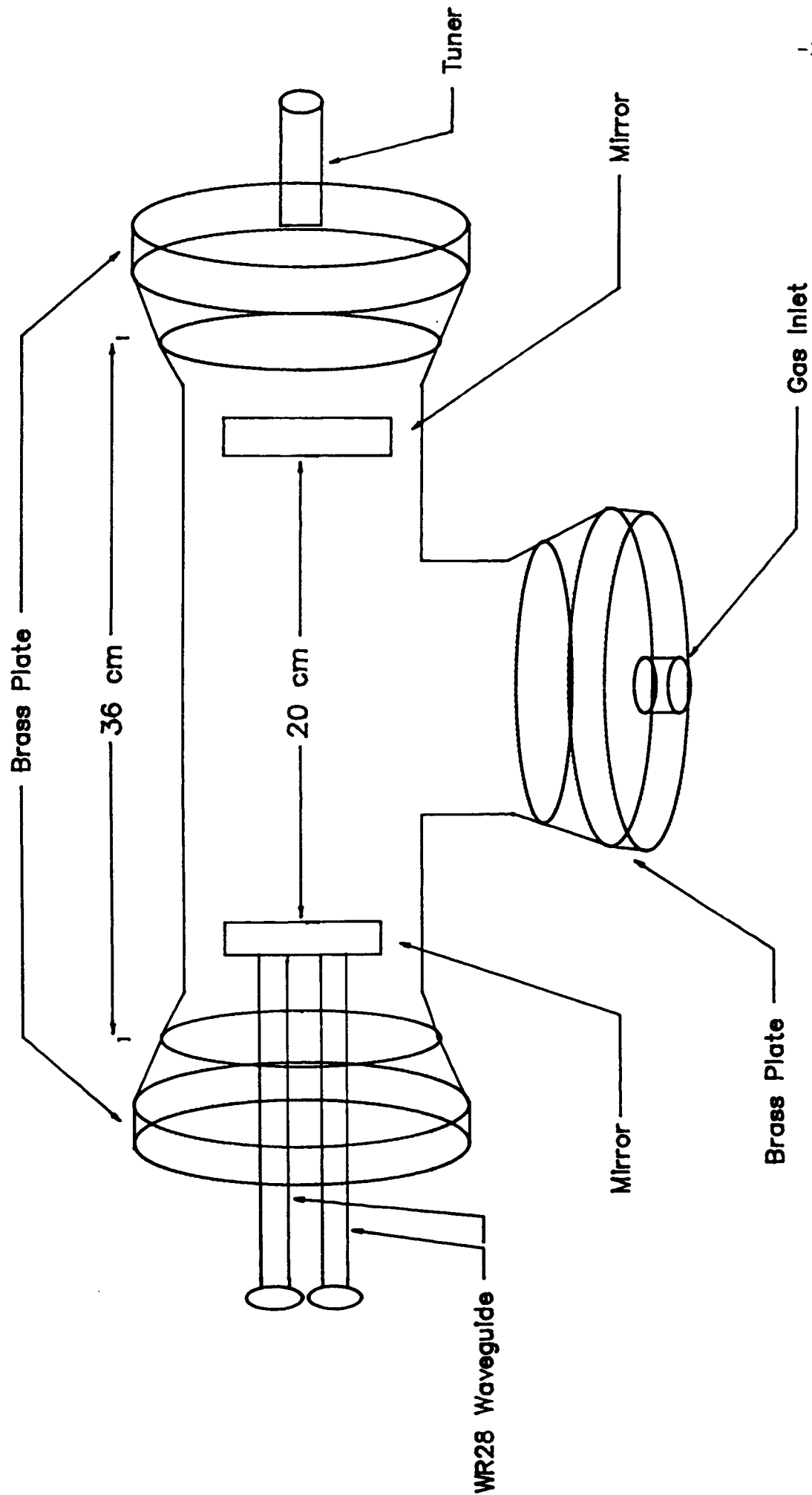
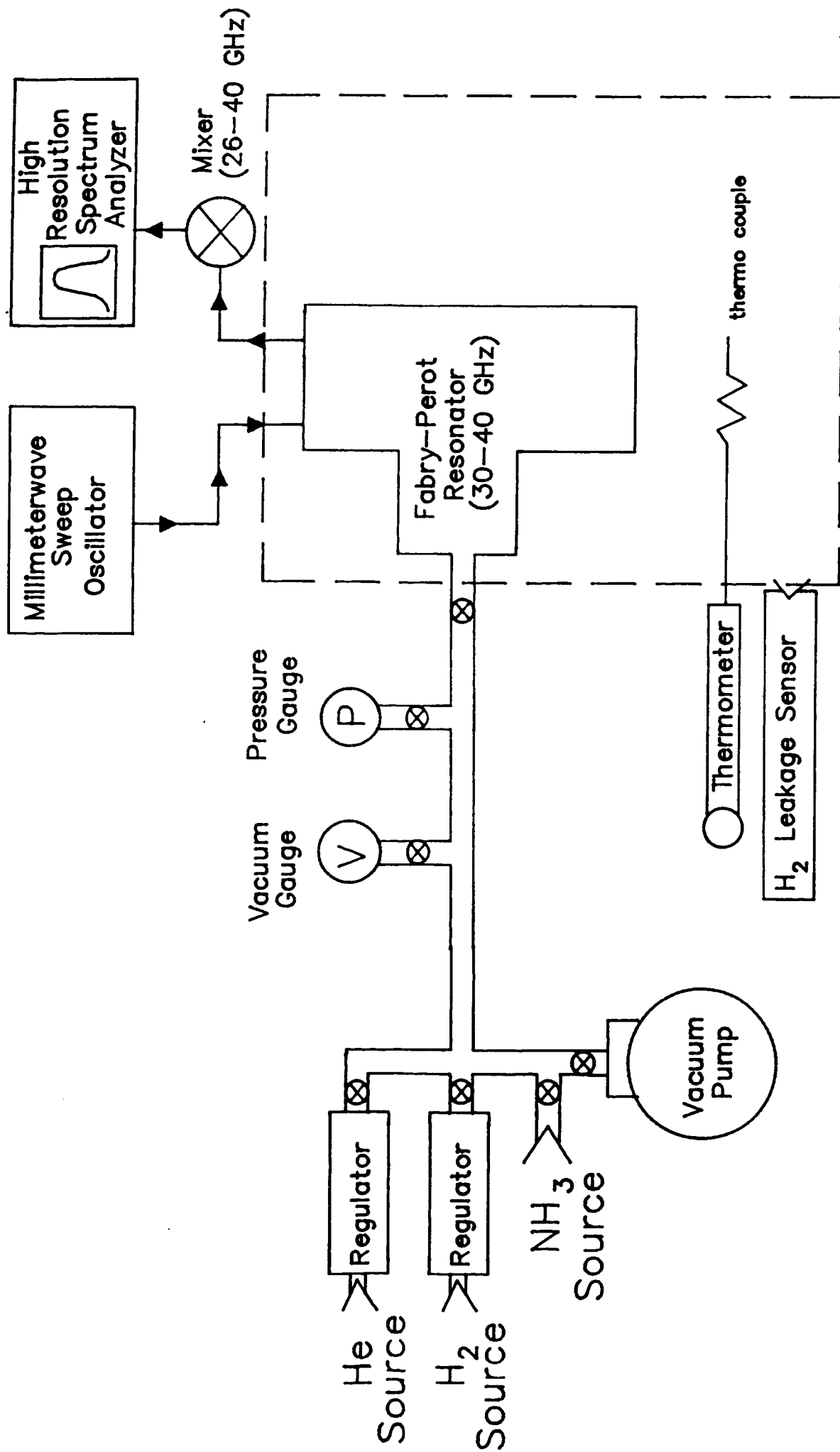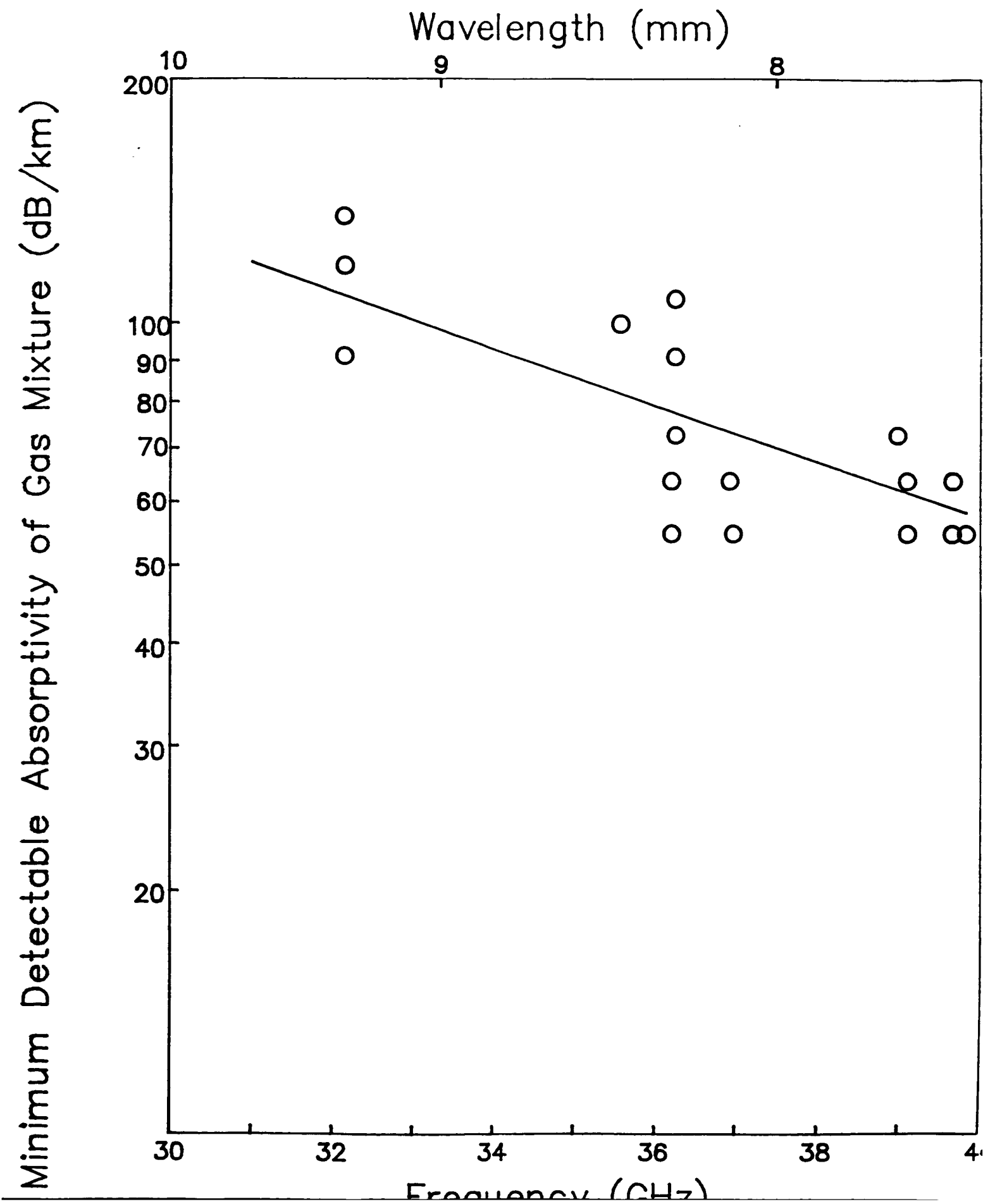# IX. KEY FIGURES

Figure 1: Diagram of Fabry-Perot Resonator

Figure 2: Block Diagram of Atmospheric Simulator (30-40 GHz)

FIGURE 4: Absorption of NH$_3$ in a 88.34% H$_2$, 9.81% He, 1.85% NH$_3$ mixture (Mixing ratio: 0.0185±0.0005) Pressure: 2 atm.   Temp: 203K
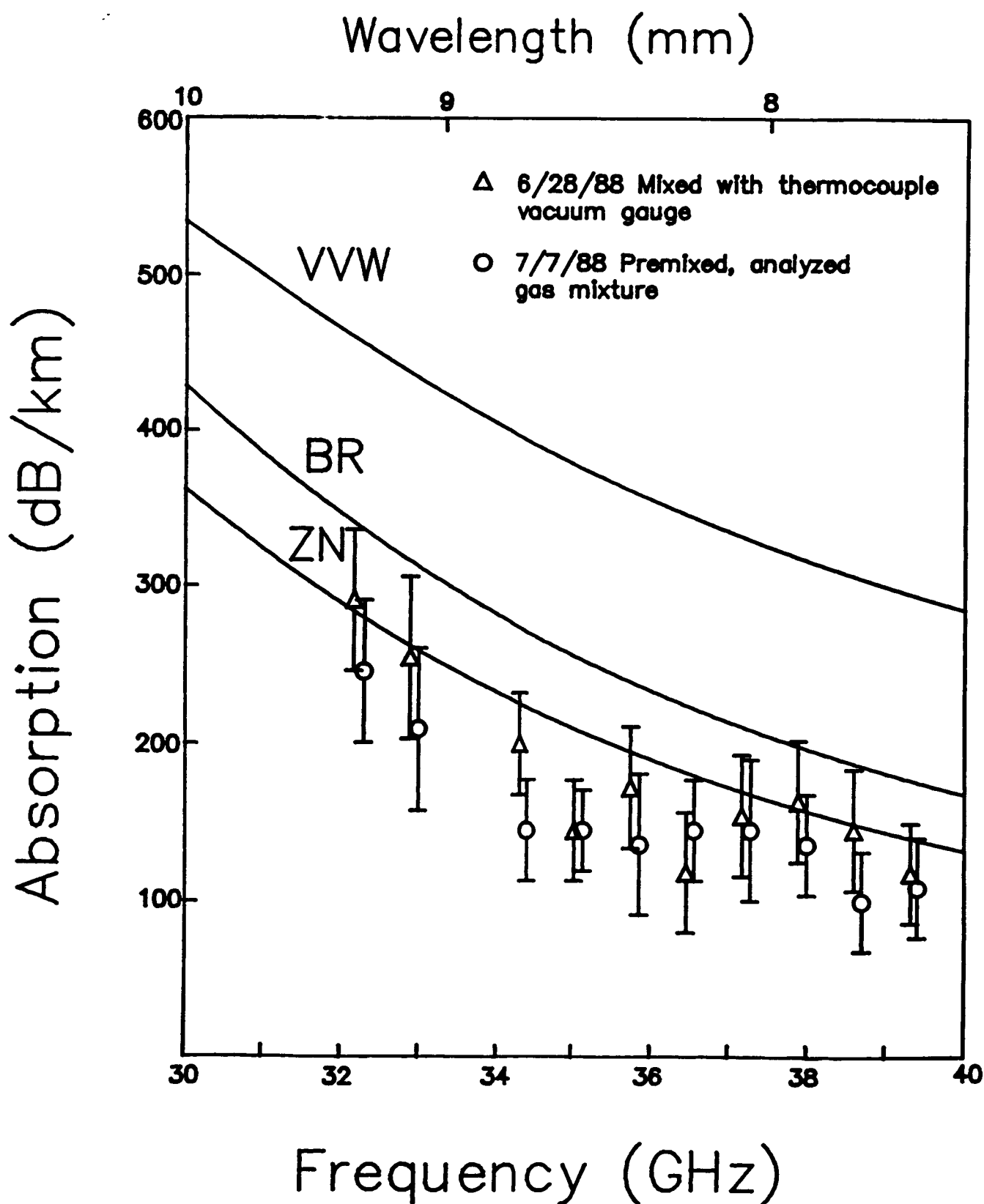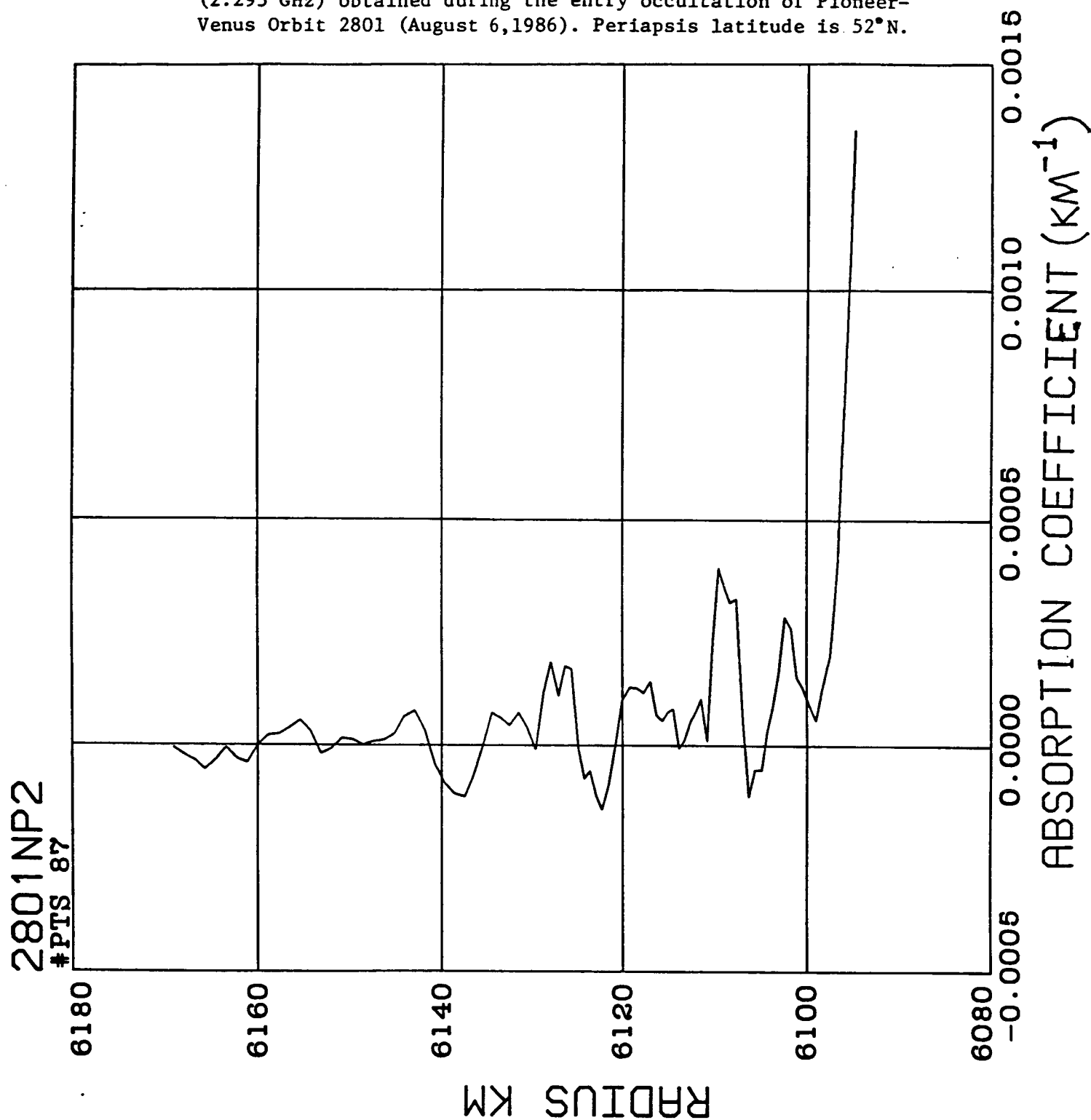

Wavelength (mm)

Figure 5: Absorptivity profile (preliminary) derived from 13-cm radio data (2.293 GHz) obtained during the entry occultation of Pioneer-Venus Orbit 2801 (August 6,1986). Periapsis latitude is 52°N.

Figure 6:   Comparison of absorptivities measured with radio occultation
technique (circular points -- from Pioneer-Venus Orbit 2801-
entry occultation) with absorptivity which would result from
saturation abundance of H_2SO_4 (from Steffes, 1985). The
absorption coefficient scale is logarithmic (exponents of 10).
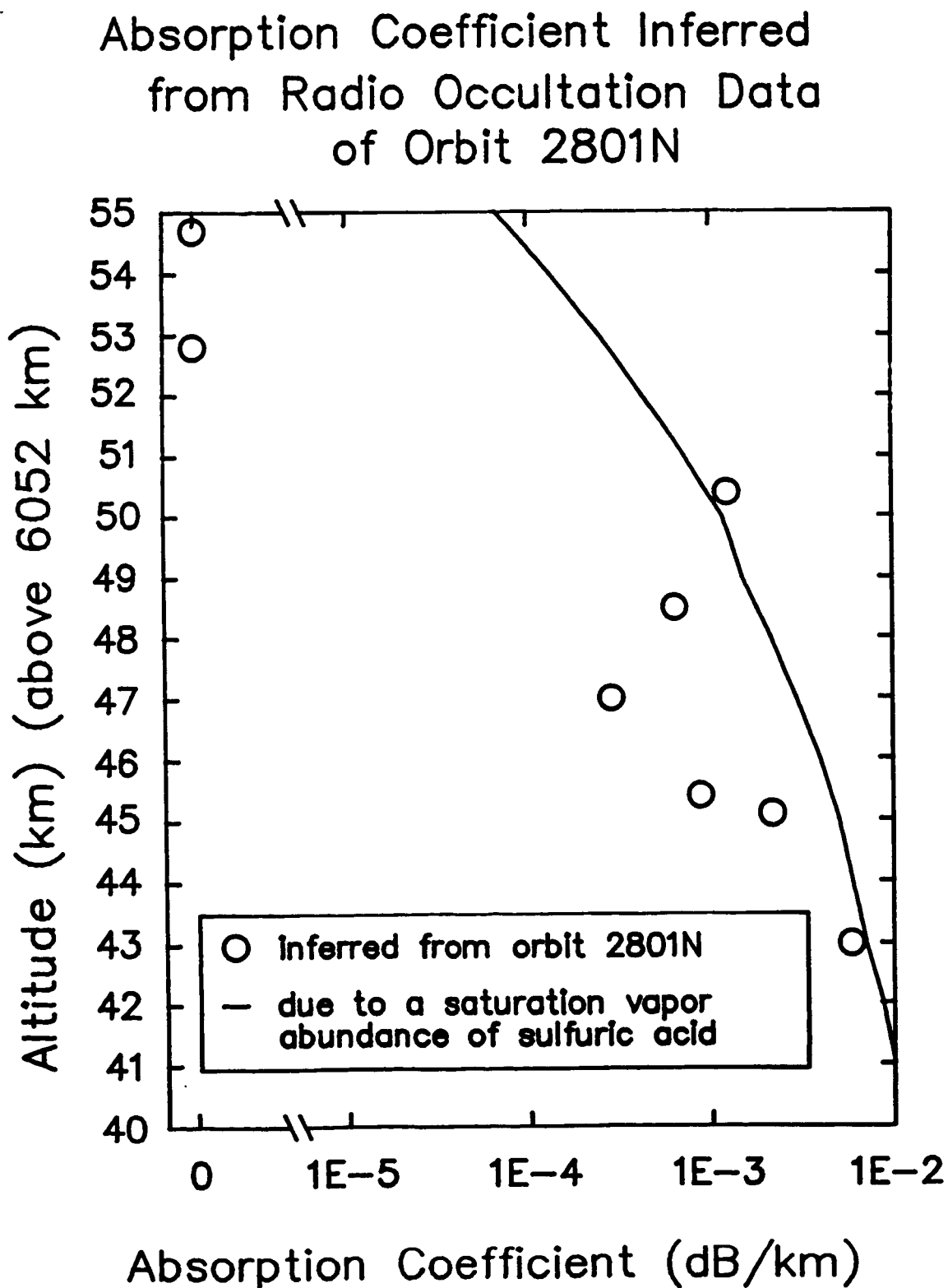All measurements were made at the 13-cm wavelength (2.293 GHz).

Absorption Coefficient Inferred
from Radio Occultation Data
of Orbit 2801N

Figure 7: Abundances of gaseous H_2SO_4 inferred from Pioneer-Venus
13-cm absorptivity profiles (circular points) compared
with the saturation abundance profile of gaseous H_2SO_4
(from Steffes, 1985). The mixing ratio scale is
logarithmic (exponents of 10).

Sulfuric Acid (H_2SO_4) Vapor Abundance

Legend:
○ inferred from orbit 2801N
— saturation vapor abundance

X-axis: Mixing Ratio (0, 1E-7, 1E-6, 1E-5, 1E-4)

Y-axis: Altitude (km) (above 6052 km)